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(54) DIELECTRIC CERAMIC COMPOSITION AND CERAMIC CAPACITOR

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(58) **Field of Search** 501/32, 138, 139; 361/321.4, 321.5

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(57) ABSTRACT

The ceramic capacitor in accordance with the present invention is fabricated by employing a dielectric ceramic composition in forming dielectric layers thereof, wherein the dielectric ceramic composition contains an oxide of Ba and Ti, an oxide of Re (Re used herein represents one or more rare-earth elements selected from Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Tb and Y), an oxide of Mg, one or more oxides selected from oxides of Mn, V and Cr, an oxide of Mo and/or W and SiO_2 or a glass component including SiO_2, wherein the amount of the oxide of Ba and Ti is 100 mol % in terms of $BaTiO_3$, the amount of the oxide of Re is 0.25 to 1.5 mol % in terms of Re_2O_3, the amount of the oxide of Mg is 0.2 to 1.5 mol % in terms of MgO and the amount of one or more oxides of Mn, V or Cr is 0.03 to 0.6 mol % in terms of Mn_2O_3, V_2O_5, Cr_2O_3, respectively, and the amount of the oxide of Mo and/or W is 0.025 to 0.25 mol % in terms of MoO_3 and WO_3.

14 Claims, 3 Drawing Sheets

FIG. 1

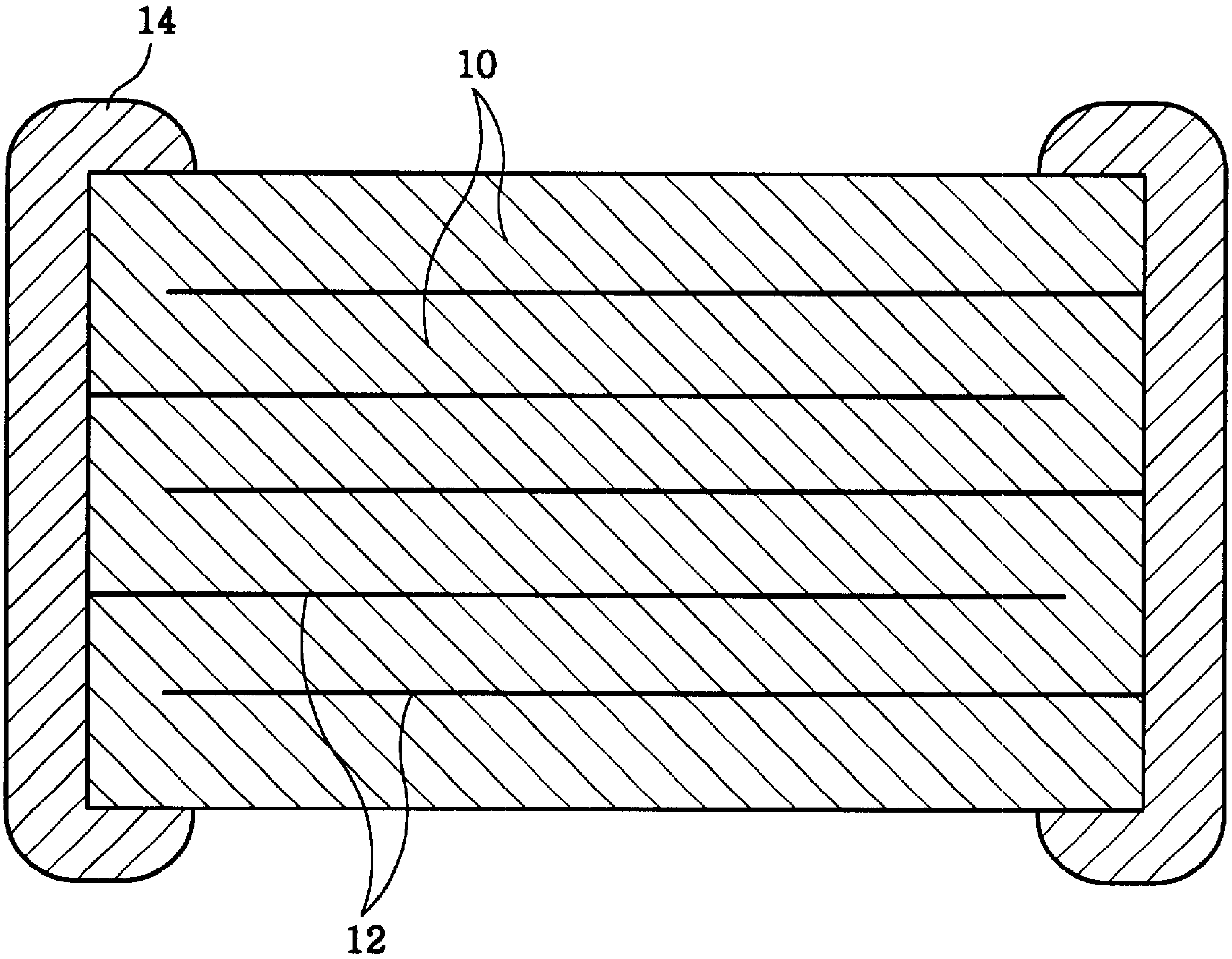


FIG.2

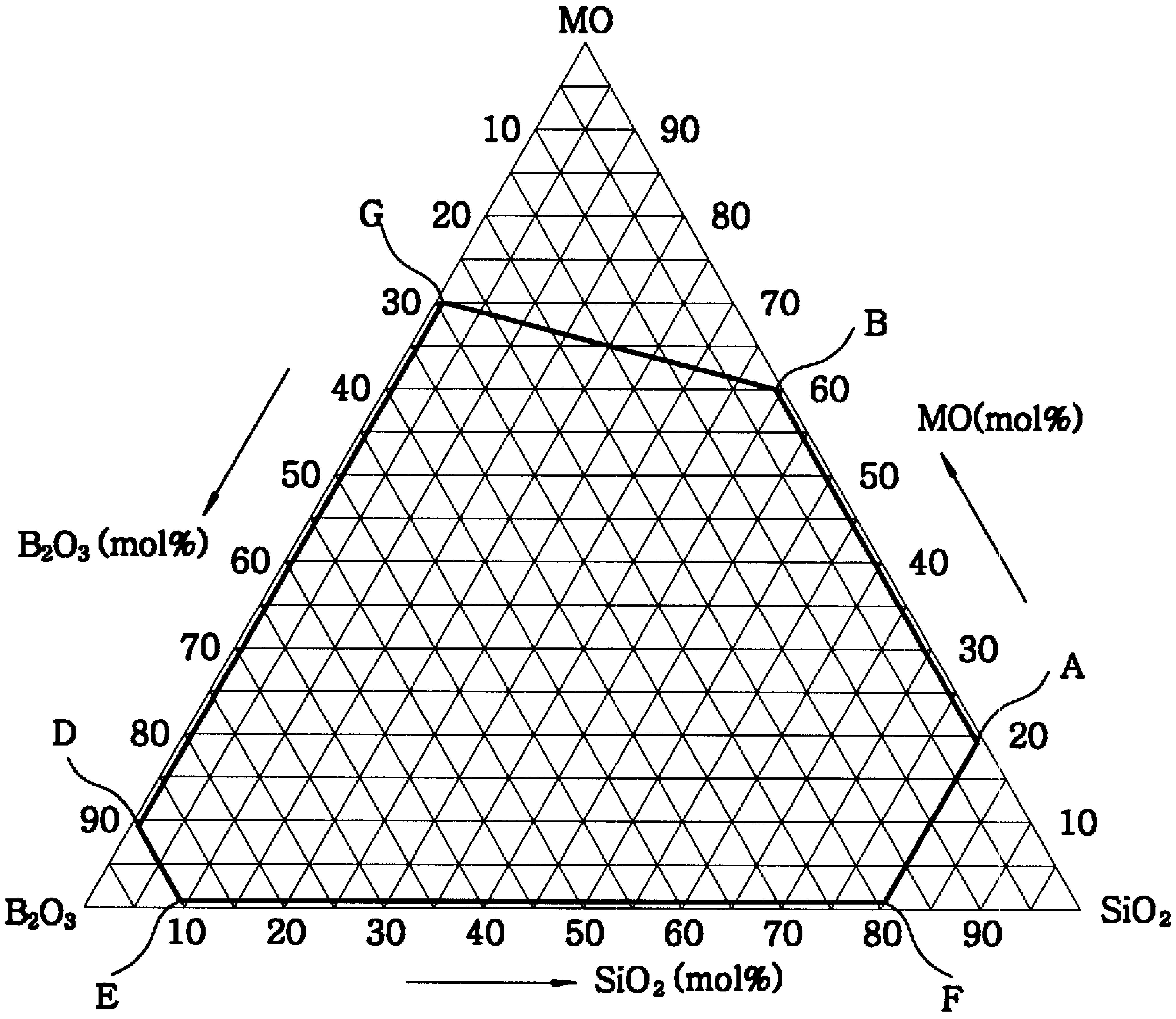
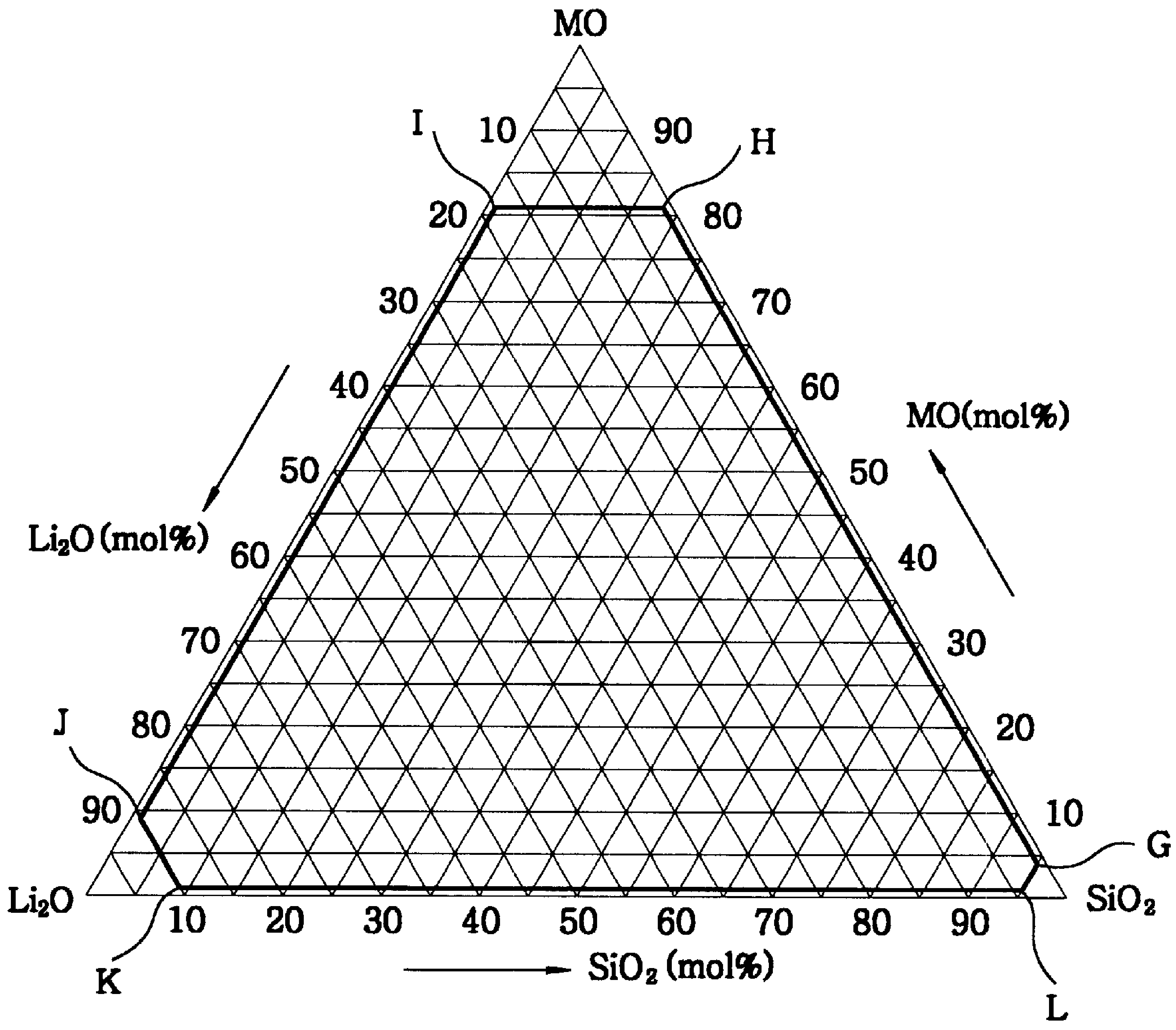

MO
10
20
30
40
50
60
70
80
90
B₂O₃ (mol%)
B₂O₃
G
D
E
90
80
70
60
50
40
30
20
10
MO(mol%)
B
A
SiO₂
10
20
30
40
50
60
70
80
90
SiO₂ (mol%)
F

FIG. 3


MO
I
H
10
90
20
80
30
70
40
60
50
50
60
40
70
30
80
20
90
10
MO(mol%)
Li₂O(mol%)
J
G
Li₂O
SiO₂
K
L
10
20
30
40
50
60
70
80
90
SiO₂(mol%)

DIELECTRIC CERAMIC COMPOSITION AND CERAMIC CAPACITOR

FIELD OF THE INVENTION

The present invention relates to a ceramic capacitor and ceramic compositions therefor; and, more particularly, to reduction resistive dielectric ceramic compositions suitable for use as a dielectric layer of a ceramic capacitor having internal electrodes made of a base metal such as Ni and a ceramic capacitor fabricated by employing such ceramic compositions as a dielectric layer thereof.

BACKGROUND OF THE INVENTION

Recently, a base metal, e.g., Ni, is widely used in forming internal electrodes of multilayer ceramic capacitors for the purpose of reducing manufacturing costs. In case the internal electrodes are composed of the base metal, it is required that chip-shaped laminated bodies including therein the internal electrodes be sintered in a reductive atmosphere in order to prevent an oxidization of the internal electrodes. Accordingly, a variety of reduction resistive dielectric ceramic compositions have been developed.

Recent trend towards ever more miniaturized and dense electric circuits intensifies a demand for a further scaled down multilayer ceramic capacitor with higher capacitance. Keeping up with such demand, there has been made an effort to fabricate thinner dielectric layers and to stack a greater number of the thus produced dielectric layers.

However, when the dielectric layers are thinned out, a voltage applied to a unit thickness intrinsically increases. Accordingly, the operating life of the dielectric layers is shortened and thus a reliability of the multilayer ceramic capacitor is also deteriorated.

SUMMARY OF THE INVENTION

It is, therefore, an object of the present invention to provide highly reliable dielectric ceramic compositions and ceramic capacitors prepared by employing such dielectric ceramic compositions in forming dielectric layers thereof, wherein the dielectric ceramic compositions exhibit such

electrical characteristics as a dielectric constant equal to or greater than 3000, a capacitance variation of −15% to +15% (based on a capacitance obtained at a temperature of +25° C.) in the temperature range from −55° C. to +125° C., a dielectric loss "tanδ" of 3.5% or less and an accelerated life of 200,000 seconds or greater.

In accordance with of the present invention, there is provided a dielectric ceramic composition comprising: 100 mol % of an oxide of Ba and Ti; 0.25 to 1.5 mol % of an oxide of Re, Re representing one or more elements selected from the group consisting of Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Y; 0.2 to 1.5 mol % of an oxide of Mg; 0.03 to 0.6 mol % of oxides of one or more elements selected from the group consisting of Mn, V and Cr; 0.025 to 0.25 mol % of oxides of one or two elements of Mo and W; and a glass component including SiO_2.

BRIEF DESCRIPTION OF THE DRAWINGS

The above and other objects and features of the present invention will become apparent from the following description of a preferred embodiment given in conjunction with the accompanying drawings, in which:

FIG. **1** represents a schematic cross sectional view illustrating a multilayer ceramic capacitor;

FIG. **2** is a triangular composition diagram for showing compositions of B_2O_3—SiO_2-MO in a unit of mol %; and

FIG. **3** sets forth a triangular composition diagram for illustrating compositions of LiO_2—SiO_2-MO in a unit of mol %.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

Compound powders of $BaCO_3$, TiO_2, Re_2O_3, MgO, Mn_2O_3, V_2O_5, Cr_2O_3, Fe_2O_3, NiO, CuO, MoO_3, WO_3 and a glass component including SiO_2 were weighed in amounts as specified in the accompanying Tables 1-1 and 1-6 and mixed for about 20 hours by a wet method in a ball mill containing therein PSZ (partially sterilized zirconia) balls and water to thereby obtain a ceramic slurry. The produced ceramic slurry was dehydrated and then dried by being heated at about 200° C. for 5 hours.

TABLE 1-1

Sample	Rare-earth (Re_2O_3)						Total		
Number	Element	Content	MgO	Mn_2O_3	V_2O_5	Cr_2O_3	Content	MoO_3	※Li_2O—
1※	Ho	1.0	0.6	0.02	—	—	0.02	0.1	0.1
2※	Ho	1.0	0.6	—	0.02	—	0.02	0.1	0.1
3※	Ho	1.0	0.6	—	—	0.02	0.02	0.1	0.1
4	Ho	1.0	0.6	0.03	—	—	0.03	0.1	0.1
5	Ho	1.0	0.6	—	0.03	—	0.03	0.1	0.1
6	Ho	1.0	0.6	—	—	0.03	0.03	0.1	0.1
7	Ho	1.0	0.6	0.01	0.02	—	0.03	0.1	0.1
8	Ho	1.0	0.6	0.05	0.02	—	0.07	0.1	0.1
9	Ho	1.0	0.6	0.05	—	0.1	0.15	0.1	0.1
10	Ho	1.0	0.6	0.05	0.01	0.1	0.16	0.1	0.1
11	Ho	1.0	0.6	0.1	0.05	0.1	0.25	0.1	0.1
12	Ho	1.0	0.6	0.1	0.1	0.1	0.3	0.1	0.1
13	Ho	1.0	0.6	0.3	—	—	0.3	0.1	0.1
14	Ho	1.0	0.6	—	—	0.3	0.3	0.1	0.1
15	Ho	1.0	0.6	—	—	0.3	0.3	0.1	0.1
16	Ho	1.0	0.6	0.6	—	—	0.6	0.1	0.1
17	Ho	1.0	0.6	—	—	0.6	0.6	0.1	0.1
18	Ho	1.0	0.6	—	—	0.6	0.6	0.1	0.1
19	Ho	1.0	0.6	0.3	0.3	—	0.6	0.1	0.1

TABLE 1-1-continued

Sample	Rare-earth (Re_2O_3)						Total		
Number	Element	Content	MgO	Mn_2O_3	V_2O_5	Cr_2O_3	Content	MoO_3	※Li_2O—
20	Ho	1.0	0.6	0.3	—	0.3	0.6	0.1	0.1
21	Ho	1.0	0.6	—	0.3	0.3	0.6	0.1	0.1
22	Ho	1.0	0.6	0.2	—	0.4	0.6	0.1	0.1
23	Ho	1.0	0.6	0.1	—	0.5	0.6	0.1	0.1
24	Ho	1.0	0.6	0.2	0.2	0.2	0.6	0.1	0.1
25※	Ho	1.0	0.6	0.7	—	—	0.7	0.1	0.1
26※	Ho	1.0	0.6	—	0.7	—	0.7	0.1	0.1
27※	Ho	1.0	0.6	—	—	0.7	0.7	0.1	0.1
28	Ho	1.0	0.6	0.2	0.1	0.4	0.7	0.1	0.1
29※	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0	0.1

※Li_2O—: Li_2O—BaO—TiO_2—SiO_2 (unit: wt %)

TABLE 1-2

Sample	Rare-earth (Re_2O_3)						Total		
Number	Element	Content	MgO	Mn_2O_3	V_2O_5	Cr_2O_3	Content	MoO_3	※Li_2O—
30	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.025	0.1
31	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.05	0.1
32	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.1	0.1
33	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.2	0.1
34※	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.3	0.1
35	Ho	1.0	0.6	0.15	0.05	—	0.2	0.1	0.1
36※	Ho	0	0.6	0.15	0.05	—	0.2	0.1	0.1
37	Ho	0.25	0.6	0.15	0.05	—	0.2	0.1	0.1
38	Ho	0.5	0.6	0.15	0.05	—	0.2	0.1	0.1
39	Ho	1.0	0.6	0.15	0.05	—	0.2	0.1	0.1
40	Ho	1.5	0.6	0.15	0.05	—	0.2	0.1	0.1
41※	Ho	2.0	0.6	0.15	0.05	—	0.2	0.1	0.1
42※	Ho	4.0	0.6	0.15	0.05	—	0.2	0.1	0.1
43	Sm	0.25	0.8	0.15	0.05	—	0.2	0.1	0.1
44	Sm	0.75	0.8	0.15	0.05	—	0.2	0.1	0.1
45	Eu	0.75	0.8	0.15	0.05	—	0.2	0.1	0.1
46	Gd	0.75	0.8	0.15	0.05	—	0.2	0.1	0.1
47	Tb	0.75	0.8	0.15	0.05	—	0.2	0.1	0.1
48	Dy	0.75	0.8	0.15	0.05	—	0.2	0.1	0.1
49	Er	0.75	0.4	0.15	0.05	—	0.2	0.1	0.1
50	Tm	0.75	0.4	0.15	0.05	—	0.2	0.1	0.1
51	Yb	0.75	0.4	0.15	0.05	—	0.2	0.1	0.1
52	Yb	1.0	0.4	0.15	0.05	—	0.2	0.1	0.1
53	Y	1.0	0.4	0.15	0.05	—	0.2	0.1	0.1
54	Ho/Dy	0.5/0.5	0.6	0.15	0.05	—	0.2	0.1	0.1
55	Ho/Dy/Yb	0.5/0.5/0.5	0.6	0.15	0.05	—	0.2	0.1	0.1
56	Sm/Ho/Yb	0.2/0.5/0.1	0.6	0.15	0.05	—	0.2	0.1	0.1
57	Sm/Yb	0.5/1.0	0.6	0.15	0.05	—	0.2	0.1	0.1
58※	Ho	1	0	0.15	0.05	—	0.2	0.1	0.1

※Li_2O—: Li_2O—BaO—TiO_2—SiO_2 (unit: wt %)

TABLE 1-3

Sample	RE (Re_2O_3)						Total			B_2O_3—MO—SiO_2				Total
No	Elmt	Cnt	MgO	Mn_2O	V_2O	Cr_2O_3	Content	MoO_3	※Li_2O—	M	B_2O_3	SiO_2	Mo	Content
59	Ho	1.0	0.2	0.15	0.05	—	0.2	0.1	0.1	—	—	—	—	—
60	Ho	1.0	1.5	0.15	0.05	—	0.2	0.1	0.1	—	—	—	—	—
61※	Ho	1.0	2.0	0.15	0.05	—	0.2	0.1	0.1	—	—	—	—	—
62※	Ho	1.0	0.6	0.15	0.05	—	0.2	0.1	0	—	—	—	—	—
63	Ho	1.0	0.6	0.15	0.05	—	0.2	0.1	0.05	—	—	—	—	—
64	Ho	1.0	0.6	0.15	0.05	—	0.2	0.1	0.5	—	—	—	—	—
65	Ho	1.0	0.6	0.15	0.05	—	0.2	0.1	1.0	—	—	—	—	—
66※	Ho	1.0	0.6	0.15	0.05	—	0.2	0.1	2.0	—	—	—	—	—
67※	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	15	65	20	0
68	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	15	65	20	0.05
69	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	15	65	20	2.00
70	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	15	65	20	5.00

TABLE 1-3-continued

Sample	RE (Re_2O_3)						Total			B_2O_3—MO—SiO_2				Total
No	Elmt	Cnt	MgO	Mn_2O	V_2O	Cr_2O_3	Content	MoO_3	※Li_2O—	M	B_2O_3	SiO_2	Mo	Content
71※	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	15	65	20	10.00
72※	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	95	4	1	1.00
73	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	90	9	1	1.00
74	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	90	1	9	1.00
75※	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	50	50	0	1.00
76	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	20	70	10	1.00
77	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	19	80	1	1.00
78	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	1	80	19	1.00
79※	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	4	95	1	1.00
80	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	1	39	60	1.00
81	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	29	1	70	1.00
82※	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	4	5	95	1.00
83	Ho	1.0	0.5	0.15	0.05	0.2	0.4	0.05	—	Ca	20	30	50	1.00

※Li_2O—: Li_2O—BaO—TiO_2—SiO_2 (unit: wt %)

TABLE 1-4

Sample	RE (Re_2O_3)							Ttl			Ttl	B_2O_3—MO—SiO_2				Ttl
No.	Elmt	Cnt	MgO	Mn_2O_3	V_2O_5	Cr_2O_3	α	Cnt	MoO_3	WO_3	Cnt	M	B_2O_3	SiO_2	Mo	Cnt
84※	Ho	1.0	0.6	0.02	—	—	0.01	0.03	0.05	0.05	0.1	Ba	15	65	20	1.00
85※	Ho	1.0	0.6	—	0.02	—	0.01	0.03	0.05	0.05	0.1	Ba	15	65	20	1.00
86※	Ho	1.0	0.6	—	—	0.02	0.01	0.03	0.05	0.05	0.1	Ba	15	65	20	1.00
87	Ho	1.0	0.6	0.03	—	—	0.01	0.04	0.05	0.05	0.1	Ca	15	65	20	1.00
88	Ho	1.0	0.6	—	0.03	—	0.01	0.04	0.05	0.05	0.1	Ca	15	65	20	1.00
89	Ho	1.0	0.6	—	—	0.03	0.01	0.04	0.05	0.05	0.1	Ca	15	65	20	1.00
90	Ho	1.0	0.6	0.01	0.02	—	0.01	0.04	0.05	0.05	0.1	Sr	15	65	20	1.00
91	Ho	1.0	0.6	0.05	0.02	—	0.01	0.08	0.05	0.05	0.1	Sr	15	65	20	1.00
92	Ho	1.0	0.6	0.05	—	0.1	0.01	0.16	0.05	0.05	0.1	Sr	15	65	20	1.00
93	Ho	1.0	0.6	0.05	0.01	0.1	0.01	0.17	0.05	0.05	0.1	Sr	15	65	20	1.00
94	Ho	1.0	0.6	0.1	0.05	0.1	0.1	0.35	0.05	0.05	0.1	Mg	15	65	20	1.00
95	Ho	1.0	0.6	0.1	0.1	0.1	0.1	0.4	0.05	0.05	0.1	Mg	15	65	20	1.00
96	Ho	1.0	0.6	0.3	—	—	0.1	0.4	0.05	0.05	0.1	Mg	15	65	20	1.00
97	Ho	1.0	0.6	—	0.3	—	0.1	0.4	0.05	0.05	0.1	Mg	15	65	20	1.00
98	Ho	1.0	0.6	—	—	0.3	0.1	0.4	0.05	0.05	0.1	Mg	15	65	20	1.00
99	Ho	1.0	0.6	0.6	—	—	0.4	1	0.05	0.05	0.1	Zn	15	85	20	1.00
100	Ho	1.0	0.6	—	0.6	—	0.4	1	0.05	0.05	0.1	Zn	35	65	20	1.00
101	Ho	1.0	0.6	—	—	0.6	0.4	1	0.05	0.05	0.1	Zn	15	65	20	1.00
102	Ho	1.0	0.6	0.3	0.3	—	0.4	1	0.05	0.05	0.1	Ba	15	65	20	1.00
103	Ho	1.0	0.6	0.3	—	0.3	0.4	1	0.05	0.05	0.1	Ba	15	65	20	1.00
104	Ho	1.0	0.6	—	0.3	0.3	0.4	1	0.05	0.05	0.1	Ba	15	65	20	1.00
105	Ho	1.0	0.6	0.2	—	0.4	0.4	1	0.05	0.05	0.1	Ba	15	65	20	1.00
106	Ho	1.0	0.6	0.1	—	0.5	0.4	1	0.05	0.05	0.1	Ba	15	65	20	1.00
107	Ho	1.0	0.6	0.2	0.2	0.2	0.4	1	0.05	0.05	0.1	Ba	15	65	20	1.00
108※	Ho	1.0	0.6	0.7	—	—	0.6	1.3	0.05	0.05	0.1	Ba/Ca	15	65	10/10	1.00
109※	Ho	1.0	0.6	—	0.7	—	0.6	1.3	0.05	0.05	0.1	Ba/Ca	15	65	10/10	1.00
110※	Ho	1.0	0.6	—	—	0.7	0.6	1.3	0.05	0.05	0.1	Ba/Ca	15	65	10/10	1.00

※ α: FeO—NiO—CuO (unit: mol %)

TABLE 1-5

Sample	Rare-earth (Re_2O_3)						Total			Total		
Number	Element	Content	MgO	Mn_2O_3	V_2O_5	Cr_2O_3	Content	MoO_3	WO_3	Content	※LiO—	SiO_2
111※	Ho	1.0	0.6	0.15	0.05	—	0.2	0.05	—	0.05	—	0.0
112	Ho	1.0	0.6	0.15	0.05	—	0.2	0.05	—	0.05	—	0.2
113	Ho	1.0	0.6	0.15	0.05	—	0.2	0.05	—	0.05	—	1.0
114	Ho	1.0	0.6	0.15	0.05	—	0.2	0.05	—	0.05	—	4.0
115※	Ho	1.0	0.6	0.15	0.05	—	0.2	0.05	—	0.05	—	5.0
116※	Ho	1.0	0.6	0.05	0.1	0.1	0.25	—	0	0	0.1	—
117	Ho	1.0	0.6	0.05	0.1	0.1	0.25	—	0.025	0.025	0.1	—
118	Ho	1.0	0.6	0.05	0.1	0.1	0.25	—	0.05	0.05	0.1	—
119	Ho	1.0	0.6	0.05	0.1	0.1	0.25	—	0.1	0.1	0.1	—
120	Ho	1.0	0.6	0.05	0.1	0.1	0.25	—	0.2	0.2	0.1	—
121	Ho	1.0	0.6	0.05	0.1	0.1	0.25	—	0.3	0.3	0.1	—

TABLE 1-5-continued

Sample	Rare-earth (Re$_2$O$_3$)						Total			Total		
Number	Element	Content	MgO	Mn$_2$O$_3$	V$_2$O$_5$	Cr$_2$O$_3$	Content	MoO$_3$	WO$_3$	Content	※LiO—	SiO$_2$
122※	Ho	1.0	0.6	0.05	0.1	0.1	0.25	—	0.4	0.4	0.1	—
123※	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0	0	0	0.1	—
124	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.01	0.01	0.025	0.1	—
125	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.02	0.02	0.04	0.1	—
126	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0	0.05	0.05	0.1	—
127	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.025	0.05	0.075	0.1	—
128	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.05	0.05	0.1	0.1	—
129	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.1	0.05	0.15	0.1	—
130	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.2	0.05	0.25	0.1	—
131※	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.3	0.05	0.35	0.1	—
132	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.05	0	0.05	0.1	—
133	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.05	0.025	0.075	0.1	—
134	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.05	0.05	0.1	0.1	—
135	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.05	0.1	0.15	0.1	—
136	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.05	0.2	0.25	0.1	—
137※	Ho	1.0	0.6	0.05	0.1	0.1	0.25	0.05	0.3	0.35	0.1	—

※Li$_2$O—: Li$_2$O—BaO—TiO$_2$—SiO$_2$ (unit wt %)

TABLE 1-6

Sample	RE (Re$_2$O$_3$)									Ttl	Li$_2$O—SiO$_2$—MO				Ttl
No.	Elmt	Cnt	MgO	Mn$_2$O$_3$	V$_2$O$_3$	α	Cnt	MoO$_3$	WO$_3$	Cnt	M	Li$_2$O	SiO$_2$	Mo	Cnt
138※	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	15	65	20	0
139	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	15	65	20	0.05
140	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	15	65	20	2
141	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	15	65	20	5
142※	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	15	65	20	10
143※	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	95	4	1	1
144	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	90	9	1	1
145	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	89	1	10	1
146※	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	50	50	0	1
147	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	20	70	10	1
148	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	5	94	1	1
149	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	1	94	5	1
150※	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	4	95	1	1
151	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	1	79	20	1
152	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	19	1	60	1
153※	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	4	5	95	1
154	Ho	1.0	0.6	0.15	0.05	0.1	0.3	0.05	0.05	0.1	Ca	20	30	50	1

※ α: FeO—NiO—CuO (unit: mol %)

Thereafter, the dried ceramic slurry was ground and then calcined in air at about 800° C. for 3 hours. The calcined slurry was then disaggregated by a wet method in a ball mill added with ethanol for about 10 hours. Next, the disaggregated ceramic slurry was dried by being heated at about 200° C. for 5 hours, thereby obtaining the powder of the calcined ceramic slurry.

In a following step, a dielectric ceramic slurry was obtained by mixing and grinding 1000 g (100 parts by weight) of the powder of the dielectric ceramic slurry, 15 wt % of an organic binder and 50 wt % of water in a ball mill, wherein the organic binder includes acrylic ester polymer, glycerin, and a solution of condensed phosphate.

Next, the dielectric slurry was subjected to a vacuum air separator to remove air bubbles therefrom and formed into a thin film coated on a polyester film by using a reverse roll coater. Thus produced ceramic thin film on the polyester film was heated and dried at about 100° C. and then diced to thereby obtain square ceramic green sheets having a thickness of about 5 μm and a size of about 10 cm×10 cm.

Meanwhile, 0.9 g of ethyl cellulose dissolved in 9.1 g of butyl carbitol and 10 g of Nickel powder having an average diameter of about 0.5 μm were loaded and stirred in a stirrer for 10 hours to form a conductive paste for use in forming internal electrodes of ceramic capacitors. Thereafter, the conductive paste was printed on the prepared ceramic green sheets to form conductive patterns thereon and then the printed conductive paste was dried.

Subsequently, ten ceramic green sheets having the conductive patterns thereon were stacked against each other with the conductive patterns facing upward, thereby forming a laminated body. Every two neighboring sheets were disposed in such a manner that the conductive patterns provided thereon were shifted by one half of a pattern size along the length direction. The laminated body also included one or more ceramic dummy sheets stacked against each of the uppermost and the lowermost ceramic green sheets having conductive patterns thereon, the ceramic dummy sheets representing ceramic green sheets without having conductive patterns thereon.

Next, the laminated body was pressed with a load of about 40 tons at about 50° C. along the stacking direction of the ceramic sheets in the laminated body. Afterwards, the pressed laminated body was diced into a multiplicity of chip shaped ceramic bodies having a size of about 3.2 mm×1.6 mm.

Thereafter, Ni external electrodes were formed at two opposite sides of each chip shaped ceramic body by, e.g., a dipping method, one end portion of each of the internal electrodes being exposed to one of the two opposite sides of each chip shaped ceramic body. Then, the chip shaped ceramic bodies were loaded into a furnace capable of controlling an atmosphere therein and the organic binder contained in the loaded ceramic bodies was removed by heating the furnace in an N_2 atmosphere. Then, the binder-removed chip shaped ceramic bodies were sintered at about 1300° C. in a non-oxidative atmosphere with oxygen partial pressure being in 10^{-5} to 10^{-8} atm order range. Thereafter, the sintered chip-shaped ceramic bodies were re-oxidized in an oxidative atmosphere to thereby obtain multilayer ceramic capacitors as shown in FIG. **1**., wherein reference numerals **10**, **12** and **14** represent dielectric layers, internal electrodes and external electrodes, respectively.

Tables 2-1 to 2-6 exhibit a measurement result of electrical characteristics obtained from the thus produced multilayer ceramic capacitors, wherein a thickness of each dielectric layer incorporated in the capacitors was about 3 μm.

The electrical characteristics of the multilayer ceramic capacitors were obtained as follows.

(A) Relative permittivity or dielectric constant ϵ_s was computed based on a facing area of a pair of neighboring internal electrodes, a thickness of a dielectric layer positioned between the pair of neighboring internal electrodes, and the capacitance of a multilayer ceramic capacitor obtained under the condition of applying at 20° C. a voltage of 1.0 V (root mean square value) with a frequency of 1 kHz.

(B) Dielectric loss tanδ (%) was obtained under the same condition as established for measuring the permittivity cited above.

(C) resistivity (Ωcm) was acquired by measuring a resistance between a pair of external electrodes after DC 25 V was applied for 60 seconds at 20° C. The number following "E" in the notation of a resistivity value presented in the accompanying Tables 2-1 to 2-6 represents an order. For instance, 4.8 E+12 represents 4.8×10^{12}.

(D) Accelerated life (second) was obtained by measuring time period until an insulation resistivity (ρ) becomes 1×10^{10} Ωcm in a DC electric field of 20 V/μm at 150° C.

(E) Capacitance variation $\Delta C/C_{25}$ (%) was obtained by measuring capacitances at −55° C., +25° C. and +125° C. in a thermostatic (or constant temperature) oven under the condition of applying a voltage of 1 V (rms value) with a frequency of 1 kHz, wherein C_{25} represents a capacitance at 25° C. and Δ C represents the difference between C_{25} and a capacitance measured at −55° C. or 125° C.

TABLE 2-1

Sample Number	Sintering Temperature (° C.)	Permittivity	Tan δ (%)	Resistivity (Ω cm) at Room Temperature	Capacitance Variation $\Delta C/C_{25}$ (%) −55° C.	125° C.	Accelerated Life (sec)
1※	1300	3400	3.3	4.8E+12	−12.0	−14.5	140,400
2※	1300	3320	3.4	9.8E+12	−13.4	−13.9	162,000
3※	1300	3680	3.6	3.1E+12	−12.5	−14.4	86,400
4	1300	3350	3.1	2.2E+12	−11.2	−13.8	244,800
5	1300	3310	3.0	1.1E+12	−11.5	−14.1	320,400
6	1300	3500	3.4	1.2E+12	−12.2	−14.5	235,400
7	1300	3440	3.3	5.5E+12	−12.1	−13.8	277,200
8	1300	3290	3.1	6.4E+12	−12.4	−13.8	295,200
9	1300	3410	3.3	7.8E+12	−12.9	−13.9	248,400
10	1300	3380	3.1	3.1E+12	−13.3	−14.1	349,200
11	1300	3150	2.8	3.1E+12	−11.2	−13.3	432,000
12	1300	3080	2.4	9.2E+11	−11.0	−14.1	560,100
13	1300	3190	2.5	3.6E+12	−12.0	−14.4	420,400
14	1300	3010	2.9	4.5E+11	−14.5	−14.1	623,800
15	1300	3620	3.5	2.7E+11	−14.8	−15.0	220,800
16	1300	3100	2.9	4.3E+12	−10.9	−12.4	1,080,400
17	1300	3030	2.4	5.5E+12	−11.3	−12.9	2,875,000
18	1300	3280	3.0	1.2E+12	−12.3	−13.5	328,900
19	1300	3080	2.6	6.5E+12	−11.5	−13.2	3,498,900
20	1300	3140	2.9	9.6E+12	−13.4	−14.3	1,094,900
21	1300	3050	2.9	3.1E+12	−13.4	−13.9	1,947,600
22	1300	3090	3.0	5.5E+12	−12.8	−13.8	335,400
23	1300	3170	3.1	2.5E+12	−10.8	−12.9	298,400
24	1300	3010	2.5	5.9E+12	−12.7	−14.8	1,048,500
25※	1300	2950	2.0	2.9E+12	−12.1	−13.9	829,000
26※	1300	2610	2.9	3.9E+11	−12.6	−14.5	1,253,400
27※	1300	2950	3.1	3.9E+11	−12.2	−15.5	145,900
28	1300	3030	2.3	3.7E+12	−11.9	−14.3	2,087,500
29※	1300	3250	3.0	4.0E+12	−13.3	−14.1	179,000

TABLE 2-2

Sample Number	Sintering Temperature (° C.)	Permittivity	Tan δ (%)	Resistivity (Ω cm) at Room Temperature	Capacitance Variation $\Delta C/C_{25}$ (%)		Accelerated Life (sec)
					−55° C.	125° C.	
30	1300	3310	3.1	3.5E+12	−13.9	−13.3	353,900
31	1300	3420	3.2	5.9E+11	−14.1	−13.3	819,400
32	1300	3140	3.4	2.2E+11	−13.9	−13.4	1,043,500
33	1300	3520	3.5	1.0E+11	−13.2	−12.8	1,567,800
34※	1300	3740	5.2	3.1E+10	−17.2	−8.2	3,255,800
35	1300	3390	3.0	5.5E+12	−13.9	−14.3	810,400
36※	1300	3980	4.4	9.2E+11	−13.9	−17.1	18,000
37	1300	3470	3.5	3.2E+12	−14.4	−14.5	221,600
38	1300	3320	3.3	3.9E+12	−13.3	−14.4	498,700
39	1300	3190	2.9	6.4E+12	−14.1	−14.5	925,800
40	1300	3040	2.8	2.2E+12	−14.9	−14.4	1,245,300
41※	1300	Incapable of obtaining a sintered ceramic with high density					
42※	1300	incapable of obtaining a sintered ceramic with high density					
43	1300	3590	3.5	2.9E+11	−14.5	−14.9	210,900
44	1300	3310	3.5	3.1E+11	−14.4	−15.0	348,000
45	1300	3190	3.2	8.1E+12	−13.3	−14.8	287,100
46	1300	3350	3.3	3.0E+12	−14.1	−14.8	453,900
47	1300	3300	3.4	3.2E+12	−14.1	−14.7	558,900
48	1300	3410	3.5	6.1E+12	−14.4	−14.9	923,400
49	1300	3090	2.8	8.2E+12	−13.7	−13.9	498,900
50	1300	3090	2.8	7.9E+12	−14.1	−13.8	348,500
51	1300	3110	2.6	3.5E+12	−14.4	−14.1	298,100
52	1300	3030	2.6	3.2E+12	−13.9	−14.2	340,400
53	1300	3350	3.2	4.1E+12	−14.4	−14.4	498,200
54	1300	3410	3.3	3.0E+12	−13.9	−14.1	598,100
55	1300	3320	3.3	2.1E+12	−14.4	−14.9	440,400
56	1300	3510	3.4	8.1E+12	−13.9	−14.9	784,300
57	1300	3280	3.3	3.9E+12	−13.4	−14.9	340,000
58※	1300	7590	8.8	4.1E+14	−45.2	12.4	285,600

TABLE 2-3

Sample Number	Sintering Temperature (° C.)	Permittivity	Tan δ (%)	Resistivity (Ω cm) at Room Temperature	Capacitance Variation $\Delta C/C_{25}$ (%)		Accelerated Life (sec)
					−55° C.	125° C.	
59	1300	3590	3.5	3.2E+12	−14.9	−13.2	697,200
60	1300	3020	3.0	3.9E+12	−13.0	−15.0	298,500
61※	1300	2950	2.2	2.1E+13	−13.1	−17.2	123,000
62※	1300	3690	4.2	4.4E+13	13.3	−15.0	12,000
63	1300	3370	3.3	9.1E+12	−13.9	−14.4	492,100
64	1300	3080	3.0	3.0E+13	−12.3	−14.1	318,000
65	1300	3010	2.5	3.1E+13	−13.0	−13.9	259,100
66※	1300	2790	2.0	4.9E+13	−13.3	−14.4	2,300
67※	1300	Incapable of obtaining a sintered ceramic with high density					
68	1300	3400	3.5	2.1E+12	−14.1	−14.5	567,800
69	1280	3290	2.5	3.6E+13	−12.5	−14.4	439,000
70	1280	3060	3.0	3.5E+13	−13.4	−13.2	650,900
71※	1280	2480	2.4	5.0E+13	−13.5	−14.1	4,500
72※	1300	Incapable of obtaining a sintered ceramic with high density					
73	1300	3290	3.4	4.4E+13	−14.4	13.9	875,600
74	1300	3350	3.5	5.3E+13	−13.5	−13.4	764,900
75※	1300	—	—	3.5E+13	—	—	
76	1300	3180	3.3	5.3E+13	−14.4	−13.3	485,900
77	1300	3080	3.2	5.9E+13	−13.1	−13.5	354,800
78	1300	3430	3.3	8.2E+13	−12.8	−15.0	298,700
79※	1300	Incapable of obtaining a sintered ceramic with high density					
80	1300	3200	3.5	3.5E+13	−14.5	−14.8	498,500
81	1300	3420	3.3	7.1E+13	−14.6	−15.0	429,800
82※	1300	Incapable of obtaining a sintered ceramic with high density					
83	1300	3310	3.5	5.7E+13	−13.8	−14.3	656,700

TABLE 2-4

Sample Number	Sintering Temperature (° C.)	Permittivity	Tan δ (%)	Resistivity (Ω cm) at Room Temperature	Capacitance Variation $\Delta C/C_{25}$ (%)		Accelerated Life (sec)
					−55° C.	125° C.	
84※	1300	3002	3.25	1.46E+12	−11.4	−11.5	29,500
85※	1300	3613	2.96	4.88E+10	−14.7	−12.1	19,700
86※	1300	3669	2.89	1.49E+12	−11.9	−13.4	138,300
87	1300	3300	2.59	9.28E+11	−14.8	−13.7	257,100
88	1300	3281	2.86	1.92E+12	−11.2	−11.6	480,200
89	1300	3707	2.50	1.99E+12	−14.9	−12.1	343,300
90	1300	3653	3.01	1.73E+12	−12.3	−14.6	337,400
91	1300	3355	2.75	8.04E+11	−11.1	−13.7	207,800
92	1300	3636	3.19	1.18E+12	−13.8	−13.6	306,600
93	1300	3013	3.20	1.96E+12	−12.8	−12.4	351,000
94	1300	3540	2.72	5.21E+11	−12.0	−12.2	300,900
95	1300	3141	2.63	1.94E+12	−11.3	−13.4	429,200
96	1300	3084	3.29	5.23E+11	−14.1	−12.3	213,200
97	1300	3402	2.55	8.61E+11	−13.1	−14.6	449,900
98	1300	3522	2.74	1.64E+12	−13.5	−12.4	263,300
99	1300	3547	3.28	6.36E+11	−13.5	−14.5	406,700
100	1300	3611	2.92	5.97E+11	−13.1	−14.9	401,800
101	1300	3105	3.32	6.00E+11	−13.6	−13.3	351,400
102	1300	3422	3.08	1.54E+12	−12.6	−13.6	206,800
103	1300	3037	2.78	1.09E+12	−12.1	−11.2	479,700
104	1300	3753	3.19	9.37E+11	−14.0	−11.7	475,400
105	1300	3214	3.20	4.03E+11	−14.8	−14.7	404,800
106	1300	3555	3.13	1.41E+12	−14.8	−11.3	228,500
107	1300	3269	2.56	1.18E+12	−14.4	−14.1	279,400
108※	1300	2386	2.95	1.29E+11	−14.7	−14.4	320,500
109※	1300	2865	2.72	2.11E+11	−13.1	−12.6	496,700
110※	1300	2187	2.76	1.53E+12	−14.2	−13.1	167,500

TABLE 2-5

Sample Number	Sintering Temperature (° C.)	Permittivity	Tan δ (%)	Resistivity (Ω cm) at Room Temperature	Capacitance Variation $\Delta C/C_{25}$ (%)		Accelerated Life (sec)
					−55° C.	125° C.	
111※	1300	Incapable of obtaining a sintered ceramic with high density					
112	1300	3490	3.5	4.3E+12	−14.5	−14.8	875,100
113	1300	3120	2.9	2.7E+13	−14.1	−14.6	547,800
114	1300	3010	2.3	1.5E+13	−13.4	−12.8	564,000
115※	1300	2690	2.8	5.3E+13	−13.5	−14.6	5,600
116※	1300	3420	3.1	5.5E+12	−13.4	−15.6	153,800
117	1300	3330	3.1	3.5E+12	−13.9	−13.3	224,900
118	1300	3410	3.3	2.8E+12	−14.1	−13.3	332,700
119	1300	3410	3.4	3.9E+11	−13.1	−13.9	983,400
120	1300	3470	3.3	1.2E+11	−13.2	−12.8	1,173,800
121	1300	3520	3.3	1.4E+11	−14.6	−11.7	2,138,000
122※	1300	3730	4.3	4.7E+10	−17.2	−9.6	3,278,000
123※	1300	3250	3.0	4.0E+12	−13.3	−14.1	179,000
124	1300	3320	3.1	5.8E+12	−13.5	−14.2	237,000
125	1300	3350	3.2	8.2E+12	−13.8	−13.8	279,000
126	1300	3410	3.3	2.8E+12	−14.1	−13.3	332,700
127	1300	3450	3.3	1.8E+12	−14.0	−13.4	402,500
128	1300	3500	3.4	9.9E+11	−13.9	−13.2	869,800
129	1300	3540	3.5	7.6E+11	−13.5	−13.1	1,115,800
130	1300	3610	3.5	8.7E+10	−13.3	−12.6	1,408,900
131※	1300	3840	6.2	5.4E+10	−18.0	−7.3	3,384,600
132	1300	3100	2.9	4.6E+12	−13.2	−14.3	132,000
133	1300	3110	3.1	5.3E+12	−13.4	−14.4	242,000
134	1300	3350	3.2	5.6E+12	−13.6	−14.2	530,000
135	1300	3420	3.4	5.6E+12	−13.9	−13.8	889,000
136	1300	3550	3.5	5.6E+12	−13.9	−13.2	1,086,000
137※	1300	3680	4.7	5.6E+12	−14.9	−10.5	2,532,000

TABLE 2-6

Sample Number	Sintering Temperature (° C.)	Permittivity	Tan δ (%)	Resistivity (Ω cm) at Room Temperature	Capacitance Variation $\Delta C/C_{25}$ (%)		Accelerated Life (sec)
					−55° C.	125° C.	
138※	1300	Incapable of obtaining a sintered ceramic with high density					
139	1300	3314	2.82	7.36E+11	−11.3	−11.0	319,400
140	1300	3678	3.17	1.20E+12	−14.3	−12.8	469,100
141	1300	3452	2.82	6.61E+11	−14.3	−11.2	425,300
142※	1300	Incapable of obtaining a sintered ceramic with high density					
143※	1300	2843	2.87	8.17E+11	−14.4	−12.8	30,900
144	1300	3387	2.54	1.16E+12	−12.8	−14.0	377,900
145	1300	3720	3.31	1.80E+12	−11.4	−14.3	309,200
146※	1300	3527	3.36	8.10E+11	−11.1	−11.9	376,500
147	1300	3706	3.18	7.88E+10	−12.4	−12.8	470,600
148	1300	3671	3.29	5.91E+11	−11.8	−14.2	433,700
149	1300	3338	2.75	3.06E+11	−13.5	−13.6	224,900
150※	1300	Incapable of obtaining a sintered ceramic with high density					
151	1300	3161	3.16	7.62E+11	−12.8	−11.4	471,800
152	1300	3765	2.89	1.57E+12	−11.9	−12.3	299,600
153※	1300	Incapable of obtaining a sintered ceramic with high density					
154	1300	3786	2.73	4.64E+11	−14.4	−13.1	330,200

As clearly seen from Tables 1-1 to 1-6 and Tables 2-1 to 2-6, multilayer ceramic capacitors with highly improved reliability having relative permittivity ϵ_s equal to or greater than 3000, capacitance variation $\Delta C/C_{25}$ within the range from −15% to +15% at temperatures ranging from −55° C. to +125° C., tan δ of 3.5% or less and accelerated life of 200,000 seconds or greater could be obtained from the above samples sintered in a non-oxidative atmosphere even at a temperature of 1300° C. or lower in accordance with the present invention.

However, samples 1 to 3, 25 to 27, 29, 34, 36, 41, 42 58, 61, 62, 66, 67, 71, 72, 75, 79, 82, 84, 85, 86, 108 to 111, 115, 116, 122, 123, 131, 137, 138, 142, 143, 146, 150, and 153 (marked with "※" at the column of sample numbers in Tables) could not satisfy the above-specified electrical characteristics. Therefore, it appears that such samples fall outside a preferable compositional range of the present invention.

The reasons why the preferable compositional range for the dielectric ceramics in accordance with the present invention should be limited to certain values will now be described. In Tables 1-1 to 1-6, the amount of oxides of Ba and Ti was 100 mol % in terms of $BaTiO_3$ (i.e., assuming Ba and Ti are in the form of $BaTiO_3$).

First, when the content of an oxide of a rare-earth element represented by Re (Re is selected, e.g., from the group consisting of Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Y) is 0 mol % in terms of Re_2O_3 (i.e., assuming the oxide of Re is in the form of Re_2O_3) as in the sample 36, the capacitance variation $\Delta C/C_{25}$ of a produced multilayer ceramic capacitor goes beyond the range from −15% to +15% when temperature varies from −55° C. to +125° C. or a desired accelerated life may not be attained; whereas when the oxide of Re is set to be 0.25 mol % in terms of Re_2O_3 as in sample 37, the desired electrical characteristics can be successfully obtained.

Further, when the content of the oxide of the rare-earth element Re is equal to or greater than 2.0 mol % in terms of Re_2O_3 as in the samples 41 and 42, highly densified ceramic bodies may not be obtained by the sintering at 1300° C.; whereas when the oxide of the rare-earth element Re is set to be 1.5 mol % in terms of Re_2O_3 as in sample 40, the desired electrical characteristics can be successfully obtained.

Accordingly, the preferable range of the content of oxide of the rare-earth element Re is from 0.25 to 1.5 mol % in terms of Re_2O_3.

It is noted that same effects can be produced regardless of whether a single rare-earth element is used as in samples 43 to 53, or two or more of rare-earth elements are used together as in samples 54 to 57 as long as the above-described preferable content range of the rare-earth element Re is satisfied.

When the content of an oxide of Mg is 0 mol % in terms of MgO as in the sample 58, the capacitance variation $\Delta C/C_{25}$ of a produced multilayer ceramic capacitor may exceed the range from −15% to +15% when the temperature varies from −55° C. to +125° C., or tanδ may be deteriorated over 3.5%; whereas when the content of the oxide of Mg is be 0.2 mol % in terms of MgO as in sample 59, the desired electrical characteristics can be successfully obtained.

In addition, when the content of the oxide of Mg is 2.0 mol % in terms of MgO as in the sample 61, the relative permittivity of the produced multilayer ceramic capacitors may become equal to or less than 3000 and the desired accelerated life cannot be obtained. However, when the content of the oxide of Mg is set to be 1.5 mol % in terms of MgO as in sample 60, the desired electrical characteristics can be successfully obtained.

Accordingly, the content of the oxide of Mg desirably ranges from 0.2 to 1.5 mol % in terms of MgO.

When the content of an oxide of each element Mn, V or Cr is 0.02 mol % in terms of Mn_2O_3, V_2O_5 or Cr_2O_3, as in the samples 1 to 3, the desired accelerated life of the produced multilayer ceramic capacitors may not be obtained; whereas when the total content of the oxides of Mn, V and Cr is set to be 0.03 mol % in terms of Mn_2O_3, V_2O_5 and Cr_2O_3, as in the samples 4 to 6, the desired characteristics can be successfully attained.

Further, when the content of an oxide of Mn, V or Cr is 0.7 mol % in terms of Mn_2O_3, V_2O_5 or Cr_2O_3, as in the samples 25 to 27, the relative permittivity of the capacitors becomes equal to or less than 3000. However, when the content of sum of the oxides of Mn, V and Cr is set to be 0.6 mol % in terms of Mn_2O_3, V_2O_5 and Cr_2O_3, as in samples 22 to 24, the desired characteristics can be successfully attained.

Accordingly, it is preferable that the total amount of oxides of Mn, V and Cr ranges from 0.03 to 0.6 mol % in terms of Mn_2O_3, V_2O_5 and Cr_2O_3.

Further, it is to be noted that same effects can be obtained regardless of whether an oxide of one of the elements Mn, V and Cr is used along as in samples 4 to 6 and 13 to 18, or two or more thereof are used together as in samples 7 to 12 and 19 to 24 as long as the total content thereof satisfies the above specified range.

When the total content of oxides of Mo and W is 0 mol % in terms of MoO_3 and WO_3, as in the samples 29, 116 and 123, the desired accelerated life of the produced multilayer ceramic capacitors cannot be obtained. However, if the total content of oxides of Mo and W is set to be 0.025 mol % in terms of MoO_3 and WO_3, respectively, as in samples 30, 117 and 124, the desired characteristics can be successfully attained.

Further, when the content of oxides of Mo and W is greater than 0.25 mol % in terms of MoO_3 and WO_3, as in the samples 34, 122 and 137, the desired accelerated life may not be obtained or the capacitance variation $\Delta C/C_{25}$ exceeds the range from −15 to +15% with the temperature varying from −55° C. to +125° C., or the $\tan\delta$ may be deteriorated over 3.5. However, when the total content of oxides is set to be 0.25 mol %, as in samples 130 and 136, the desired electrical characteristics can be successfully obtained.

Accordingly, it is preferable that the total content of the oxides of Mo and W ranges from 0.025 to 0.25 mol % in terms of MoO_3 and WO_3.

Furthermore, same effects can be obtained regardless of whether the oxides of Mo and W are used separately as in samples 30 to 33 and 117 to 121, or used together as in samples 124 to 130 and 132 to 136.

The optimum range of the glass component varies depending on the constituents thereof.

First, in case the glass component is substantially formed of SiO_2 only, the optimum content of the glass component is as follows:

When the content of SiO_2 is 0.00 mol % as in sample 111, a highly densified ceramic body may not be obtained by the sintering process at 1300° C.; whereas when the content of SiO_2 is set to be 0.2 mol % as in sample 112, the desired electrical characteristics can be successfully obtained.

Further, when the content of SiO_2 is 5.0 mol % as in sample 115, the desired accelerated life may not be obtained; whereas when the content of SiO_2 is set to be 4.0 mol % as in sample 114, the desired electrical characteristics can be obtained.

Accordingly the content of the glass component mainly formed of SiO_2 preferably ranges from 0.20 mol % and 4 mol %.

In case the glass component including SiO_2 is composed of Li_2O—BaO—TiO_2—SiO_2, the optimum range of the content of Li_2O—BaO—TiO_2—SiO_2 preferably is determined as follows:

When the total content of glass component Li_2O—BaO—TiO_2—SiO_2 is 0 mol % as in the sample 62, $\tan\delta$ of the produced capacitor may be deteriorated over 3.5% or the desired accelerated life may not be obtained; whereas when the content of the glass component Li_2O—BaO—TiO_2—SiO_2 is 0.05 mol % as in sample 63, the desired electrical characteristics can be successfully attained.

Further, when the content of the glass component Li_2O—BaO—TiO_2—SiO_2 is 2.0 mol % as in the sample 66, the relative permittivity of the produced multilayer ceramic capacitor may fall below 3000 or the desired accelerated life may not be attained; whereas when the content of the glass component Li_2O—BaO—TiO_2—SiO_2 is 1.0 mol % as in the sample 65, the desired electrical characteristics can be obtained.

Accordingly, the total content of the glass component Li_2O—BaO—TiO_2—SiO_2 is preferably between 0.05 and 1.0 wt % inclusive.

In case the glass component including SiO_2 is composed of B_2O_3—SiO_2-MO (MO used herein represents one or more oxides selected from the group of BaO, SrO, CaO, MgO and ZnO), the preferable composition of B_2O_3—SiO_2-MO for obtaining desired electrical characteristics is within the range surrounded by 6 lines formed by cyclically connecting 6 points A, B, C, D, E and F in that order shown in a triangular composition diagram of FIG. **2**, wherein the triangular composition diagram exhibits a composition of B_2O_3—SiO_2-MO in terms of their mol %. The first point A represents a composition containing 1 mol % of B_2O_3, 80 mol % of SiO_2 and 19 mol % of MO, a second point B represents a composition including 1 mol % of B_2O_3, 39 mol % of SiO_2 and 60 mol % of MO. The third point C represents a composition containing 29 mol % of B_2O_3, 1 mol % of SiO_2 and 70 mol % of MO. The fourth point D represents a composition containing 90 mol % of B_2O_3, 1 mol % of SiO_2 and 9 mol % of MO. The fifth point E represents a composition containing 90 mol % of B_2O_3, 9 mol % of SiO_2 and 1 mol % of MO and the sixth point F represents a composition containing 19 mol % of B_2O_3, 80 mol % of SiO_2 and 1 mol % of MO. If a B_2O_3—SiO_2-Mo composition is within the range defined with 6 points described above as in samples 73, 74, 76 to 78, 80, 81 and 83, the desired electrical characteristics can be obtained. However, if the composition is out of the range not as in the samples 72, 75, 79 and 82, a highly densified ceramic body may not be attained at 1300° C.

Further, when the content of B_2O_3—SiO_2-MO is 0 wt % as in the sample 67, a highly densified ceramic body may not be obtained when sintered at 1300° C.; whereas when the content of B_2O_3—SiO_2-Mo is 0.05 wt % as in sample 68, the desired electrical characteristics can be successfully attained.

Still further, when the content of B_2O_3—SiO_2-Mo is 10.00 wt % as in the sample 71, the relative permittivity may become less than 3000 or the desired accelerated life may not be obtained; whereas when the content of B_2O_3—SiO_2-Mo is set to be 5.00 wt % as in sample 70, the desired electrical characteristics can be obtained.

Accordingly, the content of B_2O_3—SiO_2-Mo preferably ranges from 0.05 to 5.0 wt %.

When the glass component including SiO_2 is composed of Li_2O—SiO_2-MO (Mo used herein represents one or more oxides selected from the group consisting of BaO, SrO, CaO, MgO and ZnO), the preferable compositional range for Li_2O—SiO_2-MO is within the range surrounded by 6 lines formed by cyclically connecting 6 points G, H, I, J, K and L in that order as shown in a triangular composition diagram of FIG. **3**, wherein the triangular diagram shows a compositional of Li_2O—SiO_2-MO in a unit of mol %. The seventh point G represents a composition containing 1 mol % of Li_2O, 94 mol % of SiO_2 and 5 mol % of MO. The eighth point H represents a composition containing 1 mol % of Li_2O, 79 mol % of SiO_2 and 20 mol % of MO. The ninth point I represents a composition containing 19 mol % of Li_2O, 1 mol % of SiO_2 and 80 mol % of MO. The tenth point

J represents a composition containing 89 mol % of Li_2O, 1 mol % of SiO_2 and 10 mol % of MO. The eleventh point K represents a composition containing 90 mol % of Li_2O_3, 9 mol % of SiO_2 and 1 mol % of MO and the twelfth point L represents a composition containing 5 mol % of Li_2O, 94 mol % of SiO_2 and 1 mol % of MO. If a Li_2O—SiO_2-Mo composition falls within the range defined by the 6 G–L, as in samples 144, 145, 147 to 149, 151, 152 and 154, the desired electrical characteristics can be obtained, but if otherwise as in samples 143, 146, 150 and 153, a highly densified ceramic body with a highly improved density may not be attained after being sintered at 1300° C.

Further, when the content of Li_2O—SiO_2-MO is 0 wt % as in the sample 138, a highly densified ceramic body may not be obtained by the sintering process at 1300° C.; whereas when the content of Li_2O—SiO_2-MO is set as 0.05 wt % as in sample 139, the desired electrical characteristics can be acquired.

Still further, when the content of Li_2O—SiO_2-MO is 10.00 wt % as in the sample 142, a highly densified ceramic body may not be gained by the sintering at 1300° C.; whereas when the content of Li_2O—SiO_2-MO is set to be 5.00 wt % as in sample 141, the desired electrical characteristics can be successfully obtained.

Accordingly, the content of Li_2O—SiO_2-MO optimally ranges from 0.05 to 5.0 wt %.

Further, when the total content of oxides of Fe, Ni and Cu and oxides of Mn, V and Cr is 0.03 mol % in terms of FeO, NiO, CuO, Mn_2O_3, V_2O_5 and Cr_2O_3 as in the samples 84 to 86, the desired accelerated life may not be obtained; whereas when the total content thereof is 0.04 mol % as in samples 87 to 89, the desired electrical characteristics can be successfully obtained.

Further, when the total content of oxides of Fe, Ni and Cu and oxides of Mn, V and Cr is 1.3 mol % in terms of FeO, NiO, CuO, Mn_2O_3, V_2O_5 and Cr_2O_3, as in the samples 108 to 110, the relative permittivity of produced multilayer ceramic capacitors may go below 3000 or the desired accelerated life may not be attained; whereas when the total content is 1.00 mol % as in samples 105 to 107, the desired electrical characteristics can be successfully obtained.

Accordingly, the total amount of the oxides of Fe, Ni and Cu and the oxides of Mn, V and Cr preferably range from 0.04 to 1.00 mol %.

It should be noted that other types of raw materials can be employed as source materials for obtaining the ceramic slurry. For instance, barium acetate or barium nitrate can be used instead of $BaCO_3$.

Although the present invention has been described with reference to the multilayer ceramic capacitors only, it should be apparent to those skilled in the art that the present invention can also be applied to single-layer ceramic capacitors.

The present invention can produce a multilayer ceramic capacitor capable of providing a desired accelerated life with a highly improved reliability, wherein the capacitor exhibits a relative permittivity ϵ_s of 3000 or greater, tanδ of 3.5% or less and a capacitance variation $\Delta C/C_{25}$ ranging from −15% to +15% with the temperature variances from −55° C. to +125° C.

While the invention has been shown and described with respect to the preferred embodiments, it will be understood by those skilled in the art that various changes and modifications may be made without departing from the spirit and scope of the invention as defined in the following claims.

What is claimed is:

1. A dielectric ceramic composition comprising:

100 mol % of an oxide of Ba and Ti;

0.25 to 1.5 mol % of an oxide of Re, Re representing one or more elements selected from the group consisting of Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Y;

0.2 to 1.5 mol % of an oxide of Mg;

0.03 to 0.6 mol % of oxides of one or more elements selected from the group consisting of Mn, V and Cr;

0.025 to 0.25 mol % of oxides of one or two elements of Mo and W; and

a glass component including SiO_2.

2. The dielectric ceramic composition of claim **1**, wherein a content of the oxide of Ba and Ti is calculated by assuming that the oxide of Ba and Ti is $BaTiO_3$; a content of the oxide of Re is calculated by assuming that the oxide of Re is Re_2O_3; a content of the oxide of Mg is calculated by assuming that the oxide of Mg is MgO; a content of oxides of Mn, V and Cr is calculated by assuming that the oxides of Mn, V and Cr are Mn_2O_3, V_2O_5 and Cr_2O_3, respectively; and a content of oxides of Mo and W is calculated by assuming that the oxides of Mo and W are MoO_3 and WO_3, respectively.

3. The dielectric ceramic composition of claim **2**, further comprising one or more oxides selected from the group consisting of oxides of Fe, Ni and Cu and wherein a total content of oxides of Fe, Ni, Cu, Mn, V and Cr is 0.04 to 1.0 mol %, the total content being calculated by assuming that the oxides of Fe, Ni, Cu, Mn, V and Cr are FeO, NiO, CuO, Mn_2O_3, V_2O_5 and Cr_2O_3, respectively.

4. The dielectric ceramic composition of claim **1**, wherein the glass component is composed of Li_2O—BaO—TiO_2—SiO_2 and the content thereof ranges from 0.05 to 1.0 wt %.

5. The ceramic composition of claim **1**, wherein the glass component is composed of B_2O_3—SiO_2-MO, MO representing one or more oxides selected from the group consisting of BaO, SrO, CaO, MgO and ZnO, and wherein a composition of B_2O_3—SiO_2-MO is within a range surrounded by 6 lines formed by cyclically connecting 6 points A, B, C, D, E and F in that order in a triangular composition diagram exhibiting compositional amounts of B_2O_3, SiO_2 and Mo in a unit of mol %, and wherein a point A represents a composition including 1 mol % of B_2O_3, 80 mol % of SiO_2 and 19 mol % of MO, a point B represents a composition including 1 mol % of B_2O_3, 39 mol % of SiO_2 and 60 mol % of MO, a point C represents a composition including 29 mol % of B_2O_3, 1 mol % of SiO_2 and 70 mol % of MO, a point D represents a composition including 90 mol % of B_2O_3, 1 mol % of SiO_2 and 9 mol % of MO, a point E represents a composition including 90 mol % of B_2O_3, 9 mol % of SiO_2 and 1 mol % of MO and a point F represents a composition including 19 mol % of B_2O_3, 80 mol % of SiO_2 and 1 mol % of MO, a content of the composition B_2O_3—SiO_2-MO ranging from 0.05 to 5.0 wt %.

6. The ceramic composition of claim **1**, wherein the glass component is substantially composed of SiO_2 and a content thereof is 0.20 to 4.0 mol %.

7. The dielectric ceramic composition of claim **1**, wherein the glass component is composed of Li_2O—SiO_2-MO, MO representing one or more oxides selected from the group consisting of BaO, SrO, CaO, MgO and ZnO, and wherein the composition of Li_2O—SiO_2-MO is within a range surrounded by 6 lines formed by cyclically connecting 6 points G, H, I, J, K and L in that order in a triangular composition diagram showing compositional amounts of Li_2O, SiO_2 and MO in a unit of mol %, and wherein a point G represents a composition including 1 mol % of Li_2O, 94 mol % of SiO_2 and 5 mol % of MO, a point H represents a composition including 1 mol % of Li_2O, 79 mol % of SiO_2 and 20 mol % of MO, a point I represents a composition including 19 mol % of Li_2O, 1 mol % of SiO_2 and 80 mol % of MO, a point J represents a composition including 89 mol % of Li_2O, 1 mol % of SiO_2 and 10 mol % of MO, a point K represents a composition including 90 mol % of Li_2O_3, 9 mol % of SiO_2 and 1 mol % of MO and a point L represents a composition including 5 mol % of Li_2O, 94 mol % of SiO_2 and 1 mol % of MO, a content of the composition Li_2O—SiO_2-MO ranging from 0.05 to 5.0 wt %.

8. A ceramic capacitor comprising one or more dielectric layers made of the dielectric ceramic composition of claim **1**.

9. The ceramic capacitor of claim **8**, wherein the content of the oxide of Ba and Ti is calculated by assuming that the oxide of Ba and Ti is $BaTiO_3$; a content of the oxide of Re is calculated by assuming that the oxide of Re is Re_2O_3; a content of the oxide of Mg is calculated by assuming that the oxide of Mg is MgO; a content of oxides of Mn, V and Cr is calculated by assuming that the oxides of Mn, V and Cr are Mn_2O_3, V_2O_5 and Cr_2O_3, respectively; and a content of oxides of Mo and W is calculated by assuming that the oxides of Mo and W are MoO_3 and WO_3, respectively.

10. The ceramic capacitor of claim **8**, wherein the dielectric ceramic composition further comprises one or more oxides selected from the group consisting of oxides of Fe, Ni and Cu and wherein a total content of oxides of Fe, Ni, Cu, Mn, V and Cr is 0.04 to 1.0 mol %, the total content being calculated by assuming that the oxides of Fe, Ni, Cu, Mn, V and Cr are FeO, NiO, CuO, Mn_2O_3, V_2O_5 and Cr_2O_3, respectively.

11. The ceramic capacitor of claim **8**, wherein the glass component is composed of Li_2O—BaO—TiO_2—SiO_2 and the content thereof ranges from 0.05 to 1.0 wt %.

12. The ceramic capacitor of claim **8**, wherein the glass component is composed of B_2O_3—SiO_2-MO, MO representing one or more oxides selected from the group consisting of BaO, SrO, CaO, MgO and ZnO, and wherein a composition of B_2O_3—SiO_2-MO is within a range surrounded by 6 lines formed by cyclically connecting 6 points A, B, C, D, E and F in that order in a triangular composition diagram exhibiting compositional amounts of B_2O_3, SiO_2 and Mo in a unit of mol %, and wherein a point A represents a composition including 1 mol % of B_2O_3, 80 mol % of SiO_2 and 19 mol % of MO, a point B represents a composition including 1 mol % of B_2O_3, 39 mol % of SiO_2 and 60 mol % of MO, a point C represents a composition including 29 mol % of B_2O_3, 1 mol % of SiO_2 and 70 mol % of MO, a point D represents a composition including 90 mol % of B_2O_3, 1 mol % of SiO_2 and 9 mol % of MO, a point E represents a composition including 90 mol % of B_2O_3, 9 mol % of SiO_2 and 1 mol % of MO and a point F represents a composition including 19 mol % of B_2O_3, 80 mol % of SiO_2 and 1 mol % of MO, a content of the composition B_2O_3—SiO_2-MO ranging from 0.05 to 5.0 wt %.

13. The ceramic capacitor of claim **8**, wherein the glass component is substantially composed of SiO_2 and a content thereof is 0.20 to 4.0 mol %.

14. The ceramic capacitor of claim **8**, wherein the glass component is composed of Li_2O—SiO_2-MO, MO representing one or more oxides selected from the group consisting of BaO, SrO, CaO, MgO and ZnO, and wherein the composition of Li_2O—SiO_2-MO is within a range surrounded by 6 lines formed by cyclically connecting 6 points G, H, I, J, K and L in that order in a triangular composition diagram showing compositional amounts of Li_2O, SiO_2 and MO in a unit of mol %, and wherein a point G represents a composition including 1 mol % of Li_2O, 94 mol % of SiO_2 and 5 mol % of MO, a point H represents a composition including 1 mol % of Li_2O, 79 mol % of SiO_2 and 20 mol % of MO, a point I represents a composition including 19 mol % of Li_2O, 1 mol % of SiO_2 and 80 mol % of MO, a point J represents a composition including 89 mol % of Li_2O, 1 mol % of SiO_2 and 10 mol % of MO, a point K represents a composition including 90 mol % of Li_2O_3, 9 mol % of SiO_2 and 1 mol % of MO and a point L represents a composition including 5 mol % of Li_2O, 94 mol % of SiO_2 and 1 mol % of MO, a content of the composition Li_2O—SiO_2-MO ranging from 0.05 to 5.0 wt %.

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